FORM PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE TRANSMITTAL LETTER TO THE UNITED STATES DESIGNED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NO. PHD 99-105 U.S. Application No. (if known, see 37 CFR 1.5) 09/807687
INTERNATIONAL APPLICATION NO. PCT/EP00/08053	INTERNATIONAL FILING DATE August 16, 2000	PRIORITY DATE CLAIMED August 20, 1999

TITLE OF INVENTION
PASSIVE COMPONENT WITH COMPOSITE

APPLICANT(S) FOR DO/EO/US
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Applicant(s) herewith submit to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.

2. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.

3. [ ] This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).

4. [ ] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.

5. [ ] copy of the International Application as filed (35 U.S.C. 371 (c)(2))
   a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] has been transmitted by the International Bureau.
   c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).

6. [X] A translation of the International Application into English (35 U.S.C. 371(c)(2))

7. [ ] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] have been transmitted by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [ ] have not been made and will not be made.

8. [ ] A translation of the amendment to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).

9. [X] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).

10. [ ] A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. [ ] An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98.

12. [X] An assignment document for recording. A separate cover sheet is compliance with 37 C.F.R. 3.28 and 3.31 is included.

13. [ ] A FIRST preliminary amendment.
    [ ] A SECOND OR SUBSEQUENT preliminary amendment.

14. [ ] A substitute specification.

15. [X] A change of power of attorney and/or address letter.

16. [X] Other items or information:

    0 Sheets of Drawings

    X Authorization Pursuant to 37 CFR § 1.136(a)(3) and to Charge Deposit Account

CERTIFICATE OF MAILING

[X] Express Mail Mailing Label No. EL686949003US
Date of Deposit April 16, 2001
I hereby certify that this paper and fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231.

Michael E. Marion
Typed Name

Signature

U.S. APPLICATION NO. (If known, see 37 C.F.R. 1.5) 09/807687	INTERNATIONAL APPLICATION NO. PCT/EP00/08053	ATTORNEY'S DOCKET NUMBER PHD 99-105

17 [X] The following fees are submitted:

BASIC NATIONAL FEE (37 C.F.R. 1.492(A)(1)-(5)):

		CALCULATIONS (PTO USE ONLY)
Search Report has been prepared by the EPO or JPO	$860.00	
international preliminary-examination fee paid to USPTO (37 C.F.R. 1.482)	$690.00	
No international preliminary examination fee paid to USPTO (37 C.F.R. 1.482) but international search fee paid to USPTO (37 C.F.R. 1.445(a)(2)	$750.00	
Neither international preliminary examination fee (37 C.F.R. 1.482) nor international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO	$970.00	
International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)	$ 96.00	
ENTER APPROPRIATE BASIC FEE AMOUNT =		$ 860.00

Surcharge of $130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 C.F.R. 1.492(e)).	$	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	12 - 20 =		X $ 18.00	$	
Independent claims	4 - 3 =	1	X $ 80.00	$ 80.00	
MULTIPLE DEPENDENT CLAIMS (if applicable)			+ $270.00	$ 270.00	
TOTAL OF ABOVE CALCULATIONS =				$1,210.00	
Reductions by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 C.F.R. 1.9, 1.27, 1.28)				$	
SUBTOTAL =				$1,210.00	
Processing fee of $130.00 for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 C.F.R. 1.492(f)). +				$	
TOTAL NATIONAL FEE =				$1,210.00	
Fee for recording the enclosed assignment (37 C.F.R. 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28,3.31). $40.00 per property +				$ 40.00	
TOTAL FEES ENCLOSED =				$1,250.00	
				Amount to be refunded	$
				charged	$

a. [ ] A check in the amount $_____ to cover the above fees is enclosed.

b. [X] Please charge my Deposit Account No. 14-1270 in the amount of $1,250.00 to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fee, with the exception of the Base Issue Fee, which may be required, or credit any overpayment to Deposit Account No. 14-1270. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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(SIGNATURE)

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Passive component with composite

The invention relates to a passive component, in particular a multilayer component, comprising a dielectric and at least one electrode.

Many components having a multilayer structure are manufactured in the electronics industry. Among them are, for example, multilayer capacitors, antennas, actuators, and varistors. Multilayer capacitors are manufactured in the greatest quantities.

Ceramic multilayer components are usually manufactured in that green ceramic substrate foils alternating with layers of a metal paste for the inner electrodes are stacked on components, which are usually dielectric, whereupon the stack of ceramic and metal layers is sintered.

The quality of a multilayer component is determined both by the chemical composition of the materials used for the dielectric and for the electrodes and by the manufacturing conditions. It is first and foremost the sintering conditions which play a part in the manufacturing conditions. Various, mutually opposed oxidation and reduction reactions may take place during sintering in dependence on the sintering atmosphere. Thus, for example, barium titanate and its derivatives becomes semiconducting during sintering in a reducing atmosphere. They are unsuitable as dielectrics in this state. Sintering of the multilayer capacitors under oxidizing conditions can take place only if the electrode material consists of rhodium, palladium, or platinum. Rhodium and platinum, however, are very expensive, their cost price may account for up to 50% of the total. The development rather tends towards the use of the much cheaper metals such as Ni, Cu, Ag, or alloys thereof instead of rhodium and platinum. These metals, however, oxidize when sintered under oxidizing conditions.

There is a particular demand for ceramic materials which can be used in combination with cheaper metals, such as nickel, copper, or silver, as temperature-stable NP0 materials, which are widely used in the field of telecommunication. The use of silver electrodes is especially attractive because the internal resistance has an increasing influence at the frequencies of above 500 MHz used in telecommunication because of the so-called skin effect. It is only electrodes of copper, silver, gold, and aluminum which have sufficiently low values for the internal resistance which render possible a use at higher frequencies.

Capacitors with low, but accurate capacitance values are particularly interesting for these applications.

A reduction of the sintering temperature to below 900 °C, which renders possible the use of silver electrodes, is problematic because the dielectric materials with high melting points are to be sintered with highly reactive glasses or other compounds of low melting points. This in its turn may lead to reactions between the various phases, which will then change the temperature specification of the dielectric materials used.

The invention has for its object to provide an electronic component, in particular a multilayer component, which can be inexpensively manufactured.

This object is achieved by means of an electronic component with a dielectric and at least two electrodes, characterized in that the dielectric comprises a composite consisting of a dielectric ceramic material and an organic polymer.

Usually, passive ceramic components are sintered for strengthening the dielectric. This leads to a shrinkage, i.e. a density increase in the dielectric which will be very different in dependence on the nature of the material, the particle size distribution of the basic powder, and the reaction conditions (sintering temperature, sintering time, sintering atmosphere). In the component according to the invention, the strengthening of the dielectric is achieved by means of a polymer. The dielectric ceramic material is for this purpose first mixed with the monomer of a suitable polymer, whereupon the monomer is polymerized. Sintering is rendered unnecessary thereby.

Preferably, the organic polymer is insoluble in water.

The use of a water-insoluble polymer prevents changes in the properties and shape of the passive component and/or the dielectric which could be caused by the penetration of moisture.

It is furthermore preferred that the polymer comprises a polyimide, polyethylene, polycarbonate, or polyurethane.

These polymers wet the dielectric ceramic material and are all insoluble in water.

It may be preferred that the dielectric ceramic material has a low temperature coefficient.

Electronic components, in particular capacitors and antennas, whose dielectric has a low temperature coefficient are widely used in the field of telecommunications. The temperature specification of the dielectric is not changed because of the low temperatures in the manufacture of the passive component.

It is particularly favorable when the electrodes comprise Ag, Au, Cu, Al, or alloys of these metals.

Since sintering at high temperatures is not necessary, inexpensive metals which would be oxidized under the usual sintering conditions can be used as the electrode material. In addition, these metals have a low effective series resistance.

It is preferred that the electronic component is chosen from the group comprising capacitors, antennas, actuators, and varistors.

The invention further relates to a method of manufacturing an electronic component with a dielectric and at least two electrodes, in which method

- the dielectric ceramic material and a monomer of a polymer are mixed together,
- the mass obtained is formed,
- the monomer is partly or completely polymerized, and
- the electrodes are provided.

It may be preferred that a second polymerization step is carried out after the electrodes have been provided.

A typical manufacturing process for a multilayer component with printed metal layers comprises the following steps:

1. manufacture of a suspension from a ceramic powder, a solvent, a dispersing agent, a binder, a liquefier, etc.,
2. drawing out of the suspension into layers,
3. drying of the layers so as to obtain green ceramic foils,
4. printing of the green ceramic foils with a structured metal layer,
5. stacking of the foils,
6. laminating the stack,
7. separating into individual green products,
8. driving out of the binder by heating,
9. sintering,
10. providing the external contact paste and baking of the outer contacts.

The method according to the invention renders most of the above steps, in particular the sintering step, of this manufacturing process unnecessary. Not only does this simplify and shorten the method, it also reduces the cost.

It is preferred in all embodiments that the polymerization is thermally initiated.

The polymerization of the monomer is thermally initiated by means of temperatures below 400 °C. These low temperatures during manufacture on the one hand lead to a product of stable shape, while on the other hand the manufacturing cost and the CO_2 emission are reduced.

It is furthermore preferred that the quantity m of monomer used lies between 3% by weight ≤ m ≤ 20% by weight in relation to the quantity of dielectric ceramic material used.

The dielectric constant ε can be adjusted to a desired value by means of the mixing ratio of dielectric ceramic material and polymer in the composite material.

The invention also relates to a dielectric ceramic compound which comprises a composite of a dielectric ceramic material and an organic polymer.

A passive component of stable shape which is not limited to a planar geometry, such as, for example, a dielectric rod antenna, may be formed with the use of a dielectric ceramic compound which comprises a composite of a dielectric ceramic material and an organic polymer, and by means of the manufacturing method according to the invention.

The invention further relates to a filter arrangement with an electronic component which comprises a dielectric and at least two electrodes, wherein the dielectric comprises a composite of a dielectric ceramic material and an organic polymer.

The invention will now be explained in more detail below with reference to three embodiments.

To manufacture an electronic component according to the invention, a dielectric ceramic material is mixed with a monomer of an organic polymer which is insoluble in water. Mixing takes place preferably in an organic solvent such as, for example, a hydrocarbon, an aromatic hydrocarbon, THF, N-methylpyrrolidone, or γ-butyrolactone. The dielectric ceramic material used may be, for example, a ferroelectric material such as barium titanate, lead-zirconium-titanium oxide, or, for example, an NP0 material such as a substituted barium-neodymium-titanium perowskite with a defective structure, $(Mg,Ca)TiO_3$, $BaZrO_3$, $BaTi_4O_9$, $Ca(Zr,Ti)O_3$, or $BaO\text{-}Sm_2O_3\text{-}5TiO_2$. The monomers used may be, for example, the monomers of a polyimide, a polyethylene, a polycarbonate, or a polyurethane. The quantity m of monomer used lies between 3 and 20% by weight in relation to the quantity of dielectric ceramic material used. After the milled dielectric ceramic material has been mixed with the monomer of one of the polymers in an organic solvent, the solvent is removed. The resulting powder is granulated in that it is pressed through a sieve with a mesh

of 300 µm, and is subsequently processed in a granulating drum into a tumbled granulate. The granulate is then molded into the desired shape under pressure. These molded products are then exposed to temperatures of up to 400 °C for the thermal initialization of the polymerization of the monomers. The polymerization may take place partly or fully at this stage. Then the electrodes are fastened, for example by means of a vapor deposition process, electroless plating, silk-screen printing, electroplating, or transfer printing. The electrodes may comprise, for example, Ag, Au, Cu, Al, or alloys of these metals.

Several of such only partly polymerized forms may be stacked one on the other and subjected to a second thermally initiated polymerization step for the manufacture of multilayer components.

Alternatively, metal strips or plates may be compressed together with the granulate in the manufacture of multilayer components. Internal electrodes are thus manufactured during the polymerization.

The electronic components manufactured may be, for example, capacitors, antennas, actuators, or varistors. One or several such electronic components may be used in a filter arrangement.

Embodiments of the invention will now be explained below, representing examples of how the invention may be realized.

Embodiment 1

To manufacture a disc capacitor, 5 g of the substituted barium-neodymium-titanium perowskite with a defective structure (# corresponds to a cation void) $\{Ba_{0.242}Sr_{0.02}Ca_{0.03}Nd_{0.232}Gd_{0.23}\#_{0.246}\}[Ti_{0.97}Nb_{0.03}]O_3$ is mixed with 16.5 mg of a 1:1 mixture of pyromellithic acid dianhydride and 4,4'-diaminodiphenylether in N-methylpyrrolidone. The solvent is subsequently evaporated. The powder thus obtained is pressed through a sieve of 300 µm mesh and is subsequently processed into a tumbled granulate in a granulating drum. The granulate is then molded into a disc with a diameter of 6 mm and a thickness of 500 µm under a pressure of 400 MPa. The disc is exposed to a temperature of 380 °C in a nitrogen atmosphere. Then Au electrodes are provided on the disc by means of chemical vapor deposition (CVD).

Embodiment 2

A disc capacitor is manufactured in the same manner as in embodiment 1. However, 26.5 mg of the mixture of pyromellithic acid dianhydride and 4.4'-diaminodiphenylether is used here.

Embodiment 3

A disc capacitor is manufactured in the same manner as in embodiment 1. However, 50.1 mg of the mixture of pyromellithic acid dianhydride and 4,4'-diaminodiphenylether is used here.

All disc capacitors had a dielectric constant ε above 20, the two disc capacitors with 3.3 and 5.3% by weight of polyimide monomer even had a dielectric constant ε above 25. In addition, all disc capacitors showed an NP0 characteristic. The insulation resistance was above $3 \cdot 10^{11}$ Ωm in all cases.

Embodiment 4

For the manufacture of a dielectric antenna, 5 g of the substituted barium-neodymium-titanium perovskite with a defective structure: $\{Ba_{0.242}Sr_{0.02}Ca_{0.03}Nd_{0.232}Gd_{0.23}\#_{0.246}\}[Ti_{0.97}Nb_{0.03}]O_3$ (in which # denotes a cation void) was mixed with 50.1 mg of a 1:1 mixture of pyromellithic acid dianhydride and 4,4'-diaminodiphenylether in *N*-methylpyrrolidone. The solvent was then evaporated. The resulting powder was pressed through a sieve of 300 µm mesh and subsequently processed into a rolling granulate in a granulating drum. The granulate was then processed in a cuboid of 2 x 10 x 16 mm^3 under a pressure of 400 MPa. The cuboid was subjected to a temperature of 380° C in a nitrogen atmosphere. Then a Cu electrode covering four sides of the cuboid was vapor-deposited on a Ni/Cr adhesion layer an electrochemically reinforced to a thickness of 15 µm.

The antenna showed a resonance frequency of 900 MHz, an adaptation at the resonance of –30 dB, and an efficiency of 80%.

CLAIMS:

1. An electronic component with a dielectric and at least one electrode, characterized in that the dielectric comprises a composite consisting of a dielectric ceramic material and an organic polymer.

2. An electronic component as claimed in claim 1, characterized in that the organic polymer is insoluble in water.

3. An electronic component as claimed in claim 1, characterized in that the polymer comprises a polyimide, polyethylene, polycarbonate, or polyurethane.

4. An electronic component as claimed in claim 1, characterized in that the dielectric ceramic material has a low temperature coefficient.

5. An electronic component as claimed in claim 1, characterized in that the electrodes comprise Ag, Au, Cu, Al, or alloys of these metals.

6. An electronic component as claimed in claim 1, characterized in that the electronic component is chosen from the group comprising capacitors, antennas, actuators, and varistors.

7. A method of manufacturing an electronic component with a dielectric and at least two electrodes, which method is characterized in that

- the dielectric ceramic material and a monomer of a polymer are mixed together,
- the mass obtained is formed,
- the monomer is partly or completely polymerized, and
- the electrodes are provided.

8. A method as claimed in claim 7, characterized in that a second polymerization step is carried out after the electrodes have been provided.

9. A method as claimed in claims 7 and 8, characterized in that the polymerization is thermally initiated.

10. A method as claimed in claims 7 and 8, characterized in that the quantity m of monomer used lies between 3% by weight $\leq m \leq$ 20% by weight in relation to the quantity of dielectric ceramic material used.

11. A dielectric ceramic compound, characterized in that it comprises a composite of a dielectric ceramic material and an organic polymer.

12. A filter arrangement with an electronic component which comprises a dielectric and at least two electrodes, characterized in that the dielectric comprises a composite of a dielectric ceramic material and an organic polymer.

ABSTRACT:

The invention describes an electronic component, in particular a multilayer component, with a dielectric and at least one electrode. The dielectric comprises a composite made of a dielectric ceramic material and an organic polymer. To manufacture the electronic component, the dielectric ceramic material is mixed with a suitable monomer, the mass is formed, and the monomer is polymerized. Ceramic bodies of stable shape are obtained which can be processed further into capacitors, antennas, or other passive components in that electrodes are provided. Sintering of the electronic components is no longer necessary.

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (includes Reference to PCT International Applications)	ATTORNEY'S DOCKET NUMBER **PHD 99.105 US**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: **"Electronic composite material component"**
the specification of which (check only one item below):

☐ is attached hereto.

☐ was filed as United States application

Serial No _____

on _____

and was amended

on _____

☒ was filed as PCT international application

Number PCT/EP00/08053

on 16 August 2000

and was amended under PCT Article 19

on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:

COUNTRY	APPLICATION NUMBER	DATE OF FILING DAY, MONTH, YEAR	PRIORITY CLAIMED UNDER 35 USC 119
Germany	**19939483.0**	**20 August 1999**	**YES**

Combined Declaration For Patent Application and Power of Attorney (Continued) (includes Reference to PCT International Applications)	Attorneys Docket Number **PHD 99.105 US**

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) abnd/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (List name and registration number)

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	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE & ZIP CODE/COUNTRY

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true: and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 if Title 18 of the United states Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

SIGNATURE OF INVENTOR 201 CITY	SIGNATURE OF INVENTOR 202	SIGNATURE OF INVENTOR 203
DATE 15 March 2001	DATE 15 March 2001	DATE 15 March 2001
SIGNATURE OF INVENTOR 204	SIGNATURE OF INVENTOR 205	
DATE	DATE	